

Z_2 -vortex order of frustrated Heisenberg antiferromagnets in two dimensions

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Abstract. We discuss the recent experimental data on various frustrated quasi-two-dimensional Heisenberg antiferromagnets from the viewpoint of the Z_2 -vortex order, which include $S=3/2$ triangular-lattice antiferromagnet NaCrO_2 , $S=1$ triangular-lattice antiferromagnet NiGa_2S_4 , $S=1/2$ organic triangular-lattice antiferromagnets κ -(BEDT-TTF) $_2\text{Cu}_2(\text{CN})_3$ and $\text{EtMe}_3\text{Sb}[\text{Pd}(\text{dmit})_2]_2$, and $S=1/2$ kagome-lattice antiferromagnet volborthite $\text{Cu}_3\text{V}_2\text{O}_7(\text{OH})_2 \cdot 2\text{H}_2\text{O}$, *etc.*

1. Introduction

Recently, geometrically frustrated magnets have attracted much interest because of their unconventional ordering behaviors, including the possible quantum spin-liquid state [1, 2, 3] and the novel ordered states like chiral ordered state [4, 5, 6, 7]. Such novel ordered states as well as the associated phase transitions are often borne by novel excitations inherent to frustrated systems. A Z_2 -vortex, stabilized in a class of two-dimensional (2D) Heisenberg magnets with the locally noncollinear spin order [8], is a typical example of such novel excitations inherent to geometrically frustrated Heisenberg magnets. It was first identified in 1984 in the antiferromagnetic (AF) classical Heisenberg model on the 2D triangular lattice as a topologically stable point defect characterized by a two-valued topological quantum number [8]. In this Z_2 -vortex, Heisenberg spins, or more precisely chirality vectors, circulate around a vortex core making a topologically stable vortex (Fig.1), whereas whether they circulate in clockwise or counter-clockwise fashion makes no distinction topologically. In other words, the Z_2 -vortex has no characteristic winding number in sharp contrast to the standard vortex.

More recently, it was argued [9, 10, 11] that such a Z_2 -vortex excitation might be responsible for certain anomalous behaviors observed experimentally in several triangular-lattice Heisenberg AFs such as $S=3/2$ NaCrO_2 [12, 13, 14, 15], $S=1$ NiGa_2S_4 [16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26] and $S=1/2$ organic compounds, κ -(BEDT-TTF) $_2\text{Cu}_2(\text{CN})_3$ [27, 28, 29, 30, 31, 32, 33, 34, 35] or $\text{EtMe}_3\text{Sb}[\text{Pd}(\text{dmit})_2]_2$ [36, 37, 38, 39]. In these materials, a spin-liquid-like behavior without the standard magnetic long-range order (LRO) was observed, while all of these compounds exhibit a weak but clear transition-like anomaly at a finite temperature. As a possible explanation of such an experimental anomaly, a Z_2 -vortex driven topological transition was invoked [9, 10, 11]. Although relatively little attention has been paid in the literature to Z_2 -vortices compared with, *e.g.*, the standard Z -vortex realized in the two-dimensional XY model [40, 41], it might have important relevance to the ordering of a variety of frustrated quasi-2D magnets than hitherto

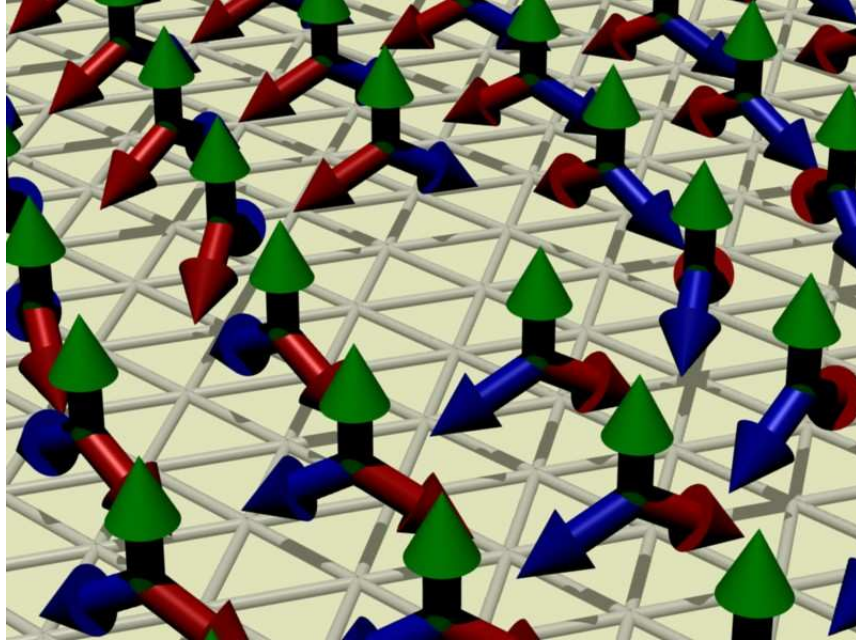


Figure 1. ‘ illustration of a Z_2 -vortex. Taken from [T. Okubo and H. Kawamura, J. Phys. Soc. Jpn. **79** (2010) 084706].

thought generally. The aim of the present article is to point out such possible relevance of Z_2 -vortex excitations to the ordering process of various geometrically frustrated magnets under recent extensive study.

It was suggested in Ref.[8] that the frustrated 2D Heisenberg AF sustaining a Z_2 -vortex might exhibit a thermodynamic phase transition at a finite temperature driven by the binding-unbinding of the Z_2 -vortices. The transition is of topological character where the ergodicity is broken topologically in the sense that the phase space is narrowed from the doubly connected one to the singly connected one. An interesting observation is that the standard AF spin correlation length ξ_s is kept finite even at and below the Z_2 -vortex transition point $T = T_v$. Nontopological excitations such as spinwaves which survive below T_v are enough to destroy the spin correlation to be paramagnetic with finite ξ_s . The low-temperature state characterized by the topologically broken ergodicity and by the finite (but often long) spin correlation length and correlation time is called a ‘spin-gel’ state [9]: See Fig.2.

Remarkable feature of the proposed Z_2 -vortex transition might be the decoupling of the two length scales: At the Z_2 -vortex transition temperature $T = T_v$, the vortex correlation length ξ_v corresponding to the mean separation of free Z_2 -vortices diverges, while the spinwave correlation length ξ_{sw} is kept finite (Fig.2(left)) [9]. Reflecting the finiteness of ξ_{sw} , the full spin correlation length ξ_s is kept finite even at $T < T_v$. Possible decoupling behaviour discussed above might be in sharp contrast to the behavior of another vortex-driven transition, *i.e.*, the standard Kosterlitz-Thouless (KT) transition of the 2D XY model, where there exists only one diverging length scale and the spin correlation length stays infinite throughout the low-temperature phase [40, 41]. Finiteness of the spin correlation length even below T_v is a pronounced feature of the spin-gel state to be contrasted to the standard KT ordered state.

Finiteness of the spin correlation length ξ_s at $T = T_v$ has important consequences on the response of the system against weak perturbative interactions coupled to the spin, *e.g.*, magnetic field, magnetic anisotropy and interplane coupling [9, 10]. In the situation where ξ_s stays

finite at the topological transition $T = T_v$, if the perturbation A is sufficiently small satisfying $A\xi_s(T = T_v)^2 < k_B T_v$, the Z_2 -vortex transition and the low-temperature ‘spin gel’ state remains essentially the same as in the unperturbed ones: See Fig.2 for comparison of the two types of vortex transitions. The situation might be contrasted to that in the KT transition of the 2D XY model, where, reflecting the divergence of the spin correlation length, even an infinitesimal perturbation leads to the 3D magnetic long-range order in real magnets.

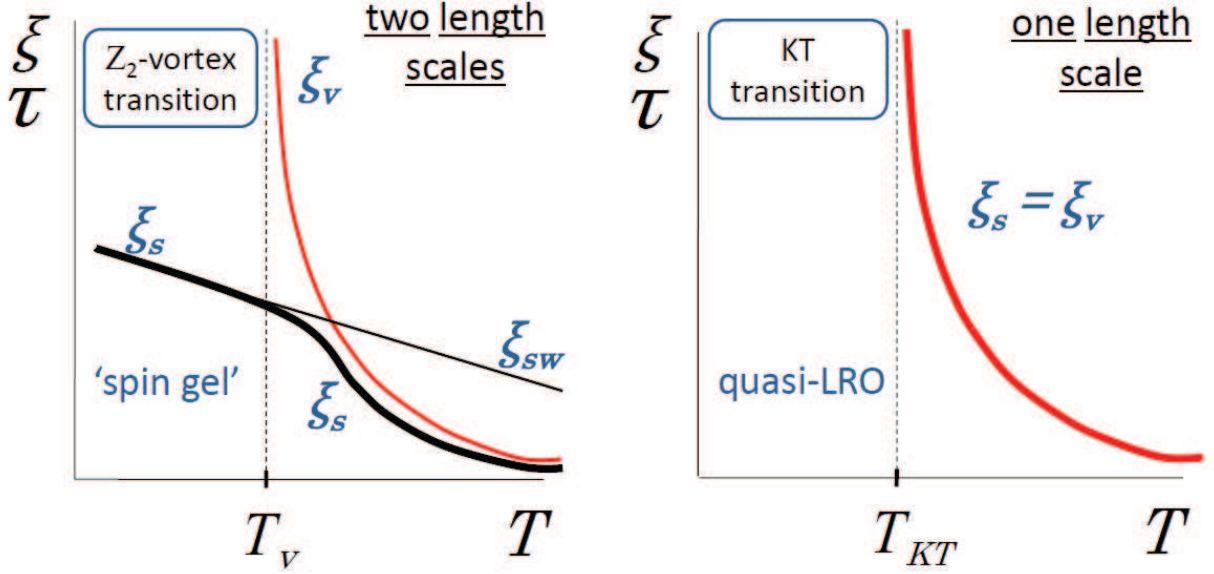


Figure 2. Schematic figure of the growth of the vortex correlation length ξ_v , the spinwave correlation length ξ_{sw} and the full spin correlation length ξ_s for each case of the Z_2 -vortex transition of the frustrated Heisenberg model (left), and of the standard Kosterlitz-Thouless transition of the two-dimensional XY model (right).

We note that the question of whether the decoupling of the two length scales, each associated with the topological and nontopological degrees of freedom, really occurs or not is still at issue. If there is no such decoupling, the vortex binding-unbinding phenomenon might eventually become a sharp crossover rather than a genuine thermodynamic transition [42]. Recent numerical simulations of the 2D triangular-lattice classical Heisenberg AF has indicated that the Z_2 -vortex binding-unbinding phenomenon looks like a thermodynamic phase transition, at least at the length scale of 10^3 lattice spacings [9]. Hence, it could be a very sharp crossover with enough experimental relevance, if not to be a genuine phase transition. In the present paper, we wish to discuss experimental consequences of such a Z_2 -vortex binding-unbinding phenomenon of the frustrated 2D Heisenberg magnets, not entering into the question of whether it is a genuine phase transition or a sharp crossover.

2. The properties of the Z_2 -vortex ordering

In this section, according to the recent theoretical analysis [9], we wish to summarize the expected properties of the Z_2 -vortex and its binding-unbinding topological transition.

- 1) When one approaches $T = T_v$ from above, the vortex correlation length ξ_v diverges as

$$\xi_v \sim \exp \left[\left(\frac{A}{T - T_v} \right)^\alpha \right], \quad T > T_v, \quad (1)$$

with $\alpha < 1$. The standard spin correlation length grows rapidly toward T_v , but remains finite even at and below $T = T_v$, exhibiting only a weak essential singularity at $T = T_v$: See Fig.2(left).

2) Singular part of the free energy around T_v is given by $f_v \approx \xi_v^{-2}$.

3) The specific heat exhibits a weak essential singularity at $T = T_v$, while a non-singular peak or a hump appears slightly above T_v . The latter corresponds to the temperature where the maximum number of Z_2 -vortex pairs dissociate giving rise to the maximum entropy change, while the transition temperature T_v corresponds to the temperature where the first Z_2 -vortex pair dissociates.

4) The magnetic susceptibility also shows a weak essential singularity at T_v .

5) The Z_2 -vortex transition accompanies an anomaly in the spin dynamics. When one approaches T_v from above, the spin correlation time grows sharply toward T_v , but not truly diverges at $T = T_v$, remaining finite even at and below T_v . Even below T_v , large and slow fluctuations should remain ('spin-gel' state).

6) The Z_2 -vortex transition and the low-temperature spin-gel state are robust against sufficiently weak magnetic perturbations such as magnetic anisotropy, interplane coupling and applied magnetic fields. When these perturbations get larger beyond a critical value, which is determined by the spin correlation length at T_v , the system would change itself into the more standard ordered states such as the AF long-range ordered state or the frozen spin-glass state, instead of the spin-gel state.

7) The dynamical spin structure $S(q, \omega)$ exhibits a characteristic central peak originated from free Z_2 -vortices above $T = T_v$ [11]. $S(q, \omega)$ also exhibits a characteristic intensity at finite ω along the zone boundary originated from Z_2 -vortex pairs.

3. Experiments on frustrated Heisenberg antiferromagnets in two dimensions

We now wish to discuss possible implications of the Z_2 -vortex ordering scenario summarized in the previous section to recent experiments on several quasi-2D frustrated Heisenberg AFs. Those include (a) $S=3/2$ triangular-lattice AF NaCrO_2 , (b) $S=1$ triangular-lattice AF NiGa_2S_4 , (c) $S=1/2$ organic triangular-lattice AFs $\kappa\text{-(BEDT-TTF)}_2\text{Cu}_2(\text{CN})_3$ and $\text{EtMe}_3\text{Sb}[\text{Pd(dmit)}_2]_2$, (d) $S=1/2$ kagome-lattice AF $\text{Cu}_3\text{V}_2\text{O}_7(\text{OH})_2 \cdot 2\text{H}_2\text{O}$, and (e) $S=3/2$ kagome-bilayer (or pyrochlore-slab) AF $\text{SrCr}_{8-x}\text{Ga}_{4+x}\text{O}_{19}$, *etc.*

(a) NaCrO_2

NaCrO_2 is a $S=3/2$ triangular-lattice Heisenberg AF with its Curie-Weiss temperature $T_{CW} \simeq 290\text{K}$. This magnet does not exhibit the conventional AF LRO down to low temperature, while it exhibits a magnetic short-range order (SRO) characterized by an incommensurate wavevector close to the 120-degrees structure [12, 13, 14, 15, 44]. The origin of the incommensurate spin structure is ascribed either to an easy-axis-type magnetic anisotropy or to a weak interplane coupling. A clear but rounded specific-heat peak is observed at $T_{peak} = 41\text{K}$, whereas a transition-like dynamical anomaly is observed at $T_f \simeq 30\text{K}$, a temperature slightly below T_{peak} . At $T = T_f$, the spin dynamics is rapidly slowed down giving rise to a quasi-static internal field. The spin dynamics probed by NMR, ESR and μSR , however, is not completely frozen below T_f . Rather, the spins remain slowly fluctuating even below T_f unlike the conventional AF or the spin glass. Such a dynamically fluctuating ordered state extends over a wide temperature range down to 10K. The spin correlation length determined from neutron scattering is kept finite $\xi \simeq 20$ lattice spacings even below T_f , which is not resolution-limited.

These experimental features are fully consistent, at least qualitatively, with the Z_2 -vortex scenario discussed in the previous section, if the experimental freezing temperature T_f is identified as T_v and the low-temperature state as the 'spin-gel' state. In particular, energetics

seems appropriate. Recent Monte Carlo simulation on the triangular-lattice classical Heisenberg model with the nearest-neighbor AF coupling yielded $T_{peak}/T_{CW} \simeq 0.137$ and $T_v/T_{CW} \simeq 0.123$ [9], which are close to the corresponding experimental values for NaCrO_2 , *i.e.*, 0.14 and 0.10, respectively. It remains to be seen whether the dynamical spin structure factor exhibits a characteristic behavior predicted as a fingerprint of the Z_2 -vortex, the point 7 above [11].

Magnetic ordering of similar chromium triangular-lattice AFs like HCrO_2 [15, 43], LiCrO_2 [15, 43, 44, 45, 46, 47, 48], CuCrO_2 [47, 49, 50, 51], AgCrO_2 [47, 52] and PdCrO_2 [53, 54] were also investigated experimentally. In contrast to NaCrO_2 , these materials exhibit a 3D AF LRO at a Neel temperature $T = T_N$ due to the weak interlayer coupling. Yet, since all these materials are magnetically quasi-2D, the Z_2 -vortex might exist in the 2D regime realized in the temperature region slightly above T_N . A particularly interesting compound in this series might be KCrO_2 , where no evidence of the 3D AF LRO was reported by neutron scattering as for NaCrO_2 [44]. Further experimental study on this compound is encouraged.

It was also reported that the two-dimensionality could strongly be enhanced in $\text{Cu}_{1-x}\text{Ag}_x\text{CrO}_2$, a compound obtained from CuCrO_2 by chemical substitution [55, 56]. Then, one might expect that the Z_2 -vortex has a higher chance to be realized in $\text{Cu}_{1-x}\text{Ag}_x\text{CrO}_2$. Of course, in this compound, care has to be taken on the possible effect of quenched randomness.

(b) NiGa_2S_4

NiGa_2S_4 is a $S=1$ triangular-lattice Heisenberg AF with its Curie-Weiss temperature $T_{CW} \simeq 80\text{K}$ [16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26]. This material attracted considerable attention of researchers, because the low-temperature specific heat exhibits a T^2 behavior suggesting the existence of Goldstone modes associated with a broken continuous symmetry, whereas no magnetic LRO was observed down to low temperature [16]. The observed magnetic SRO is an incommensurate helical structure in the basal plane close to the 60-degrees structure. Such a spin structure is stabilized by the AF third-neighbor interaction J_3 which frustrates the ferromagnetic first-neighbor coupling J_1 with $J_1/J_3 \simeq -0.2$ [23].

Meanwhile, NiGa_2S_4 , with an integer spin quantum number $S=1$ in contrast to a half-integer spin quantum number $S=3/2$ of NaCrO_2 , exhibits a strikingly similar ordering behavior to that of NaCrO_2 . Indeed, NiGa_2S_4 exhibits a clear but rounded specific-heat peak at $T_{peak} = 12\text{K}$ [23] (the peak of C/T is located at 10K), whereas a transition-like dynamical anomaly is observed at $T^* \simeq 8.5\text{K}$, a temperature slightly below T_{peak} , where the spin dynamics is rapidly slowed down. Note that quasi-static internal fields of *dipolar* nature is observed below T^* , at least at the time scale of NQR and muon measurements. The spin dynamics probed by NMR, NQR, ESR and μSR , however, is not completely frozen even below T^* . Rather, the spins remain slowly fluctuating at the time scale of MHz, unlike the conventional AF or the spin glass. Such a dynamically fluctuating ordered state extends over a wide temperature range down to 2K. The spin correlation length determined from neutron scattering is kept finite even at and below T^* , *i.e.*, $\xi \simeq 7$ lattice spacings [16]. Dynamical freezing at $T = T^*$ also accompanies a weak anomaly in the susceptibility. Anomaly in the susceptibility looks clearer than the one observed in NaCrO_2 , presumably due to the fact that NiGa_2S_4 possesses a ferromagnetic nearest-neighbor coupling. Application of magnetic fields greater than $100 \sim 1000\text{ G}$ gradually changes the slowly fluctuating state into the more conventional frozen state [20].

As for NaCrO_2 , those experimental features of NiGa_2S_4 are fully consistent, at least qualitatively, with the Z_2 -vortex ordering scenario, if the experimental freezing temperature T^* is identified as T_v and the low-temperature state as the spin-gel state. Concerning its energetics, NiGa_2S_4 is characterized by $T_{peak}/T_{CW} \simeq 0.15$ and $T_v/T_{CW} \simeq 0.11$, which are close to the corresponding theoretical values 0.133 and 0.123 [9]. Furthermore, a theoretical estimate of the crossover-field intensity $\sim 1000\text{G}$ obtained with the information of the spin correlation length $\xi \simeq 6$ lattice spacings at $T = T^*$, seems consistent with the μSR measurement [20].

In this Z_2 -vortex picture, slow dynamics below T^* is primarily borne by spinwaves. Then,

spinwaves would be responsible for the T^2 specific heat. Indeed, recent inelastic neutron-scattering measurements identified a damped spinwave excitation at a low temperature $T = 1.5\text{K}$ [25]. The T^2 behavior of the specific heat sets in around T^* , which is consistent with such a picture. Ref.[57] accounted for the T^2 specific heat based on spinwave excitations of the noncollinear AF order of $S=1$ quantum magnets, neglecting the vortex degrees of freedom [57]. Vortex-free assumption of Ref.[57] is well justified below the Z_2 -vortex transition.

Impurity effects on the low-temperature specific heat and on the transition-like anomaly T^* was studied in Ref.[24]. Upon substitution of various kinds of impurities, *i.e.*, $S = 0$ Zn^{2+} , $S = 2$ Fe^{2+} , $S = 3/2$ Co^{2+} and $S = 5/2$ Mn^{2+} , the transition-like anomaly of the pure compound at T^* gradually changed its character to that of the standard spin-glass transition in all cases studied, accompanied by a pronounced cusp and the deviation between the FC and ZFC data. By contrast, the low-temperature specific heat persistently exhibited a T^2 behavior scaled by the Curie-Weiss temperature for Zn^{2+} and Fe^{2+} with integer spins, but exhibited a different T -linear behavior characteristic of the standard spin glass for Co^{2+} and Mn^{2+} with half-integer spins. This observation seems to suggest that the nature of low-energy excitations depends significantly on the size of spins, *i.e.*, whether S being an integer or a half-integer. It remains to be seen whether the standard spinwave picture could account for such exotic properties or not. In this context, one may also notice that, in case of kagome-bilayer AF $\text{SrCr}_{8-x}\text{Ga}_{4+x}\text{O}_{19}$ (SCGO) to be discussed below, the low-temperature specific heat is observed to persistently exhibit a T^2 behavior below the spin-glass transition temperature, although the spin quantum number there is a half-integer ($S = 3/2$ for Cr^{3+}): See section 3(e) below.

Concerning the magnitude of the spin correlation length, quantitative discrepancy exists between the experimental value and the numerical value obtained for the simplest classical Heisenberg model with the nearest-neighbor AF coupling. Monte Carlo simulation yielded ξ of order 10^3 near $T = T_v$ [9], while it is only 8 or 20 experimentally. So, some mechanism, not taken into account in the simplest classical Heisenberg model, is required to explain the shortness of experimental ξ . It should be emphasized here that the magnitude of ξ is a nonuniversal property governed by the non-vortex physics, which could significantly be reduced, say, by quantum fluctuations, charge fluctuations, or further frustration effects associated with the interaction other than the main exchange couplings. Interestingly, recent ARPES study performed at higher temperatures ($100\sim 200\text{K}$) has revealed that the low-energy charge fluctuation (hole dynamics) across the Mott gap is characterized by the wavevectors which are completely different from the wavevectors characterizing the magnetic SRO at lower temperatures, the occurrence of spin-charge separation [26].

In fact, the specific heat of NiGa_2S_4 exhibits another pronounced peak at a temperature around $T \sim 100\text{K}$, an order of magnitude higher temperature than that at $T_{peak} = 10\text{K}$. This higher specific-heat peak might be related to charge fluctuations as investigated in Ref.[26]. Ref.[58] (see also Ref.[26]) suggested that the strong nematic correlations might set in around this higher specific-heat-peak temperature. However, the T -linear behavior in the specific heat mentioned above sets in at the lower specific-heat-peak temperature of 10K , not at the higher specific-heat-peak temperature of 100K , becoming vanishingly small (after the lattice contribution subtracted) in the temperature range between the two specific-heat-peak temperatures. Hence, strong nematic correlations, as are expected to accompany nematic-wave excitations giving rise to the T^2 specific heat, are unlikely to occur in the high-temperature regime of $\sim 100\text{K}$.

Finally, the noncollinear AF order might explain another noticeable feature of experiments that the T^2 specific heat is quite robust against applied magnetic fields [16]. This experimental observation is rather surprising since applied fields reduce the Hamiltonian symmetry from $O(3)$ to $O(2)$, leading to smaller number of Goldstone modes, *i.e.*, from three to one. As discussed in Ref.[59], an interesting observation here is that the noncollinear AF ground state in

magnetic fields is capable of keeping an “accidental” degeneracy not related to the Hamiltonian symmetry $O(2)$, essentially of the same amount as in the zero-field case, when the ordered state is a commensurate one, *e.g.*, the 120-degrees or the 60-degrees structure. In fact, even in applied fields, the ground-state manifold still retains *three* continuous parameters which can be set freely, just as in the zero-field case. One of these three is of symmetry origin, *i.e.* a true Goldstone mode, while other two are not of symmetry origin (accidental), *i.e.* *pseudo*-Goldstone modes. Thus, at the classical level, such pseudo-Goldstone modes may account for the robustness of the low-temperature specific heat in applied fields, while this degeneracy would become approximate in quantum systems. The degeneracy would also become approximate when the ordering wavevector deviates from the commensurate position.

(c) κ -(BEDT-TTF) $_2$ Cu $_2$ (CN) $_3$ and EtMe $_3$ Sb[Pd(dmit) $_2$] $_2$

$S=1/2$ organic triangular-lattice AFs κ -(BEDT-TTF) $_2$ Cu $_2$ (CN) $_3$ [27, 28, 29, 30, 31, 32, 33, 34, 35] and EtMe $_3$ Sb[Pd(dmit) $_2$] $_2$ [36, 37, 38, 39] exhibit a spin-liquid behavior without the conventional magnetic LRO down to low temperature. The lattice here is not a regular triangular lattice, but is a slightly distorted one. The charge and the spin densities are spread over a dimer molecule so that the system intrinsically possesses the polarization degree of freedom within a molecule. Furthermore, these materials are Mott insulators lying close to the metal-insulator boundary so that intermolecular charge fluctuations might play an important role.

Recent measurements have revealed that the specific heat [32] and the thermal conductivity [39] of these organic compounds exhibit a T -linear behavior at low temperatures, often ascribed to the fermionic ‘spinon’-like excitations [60, 61]. More precisely, for the case of κ -(BEDT-TTF) $_2$ Cu $_2$ (CN) $_3$, such a T -linear behavior was reported for the specific heat [32], while a small nonzero energy gap was reported for the thermal conductivity [33]. For EtMe $_3$ Sb[Pd(dmit) $_2$] $_2$, even the thermal conductivity exhibits a T -linear behavior [39].

Recent measurements have also revealed a weak transition-like anomaly occurring at a finite temperature just above the temperature range where the T -linear behavior is observed in the specific heat or in the thermal conductivity. In case of κ -(BEDT-TTF) $_2$ Cu $_2$ (CN) $_3$, two anomalies (or crossovers) were observed, one at ~ 6 K and the other at ~ 3 K. The higher one at 6K is characterized by the hump (or the peak) of the specific heat [32] and of the thermal expansivity coefficient [34], with a minimum of the NMR T_1^{-1} [27] and of the in-plane thermal conductivity [33]. The lower one at 3K is characterized by a weak additional structure in the specific heat [32] and in the thermal expansivity coefficient [34], with a maximum of the in-plane thermal conductivity [33]. Similar anomaly or crossover is also observed for EtMe $_3$ Sb[Pd(dmit) $_2$] $_2$ [37, 38, 39].

We note that the Z_2 -vortex order is one possible candidate of the experimentally observed anomaly. In this correspondence, one identifies the lower anomaly as the Z_2 -vortex transition temperature T_v and the higher anomaly as the specific-heat-peak temperature T_{peak} . Experimentally, the ratio T_{peak}/T_v comes around two in these organic compounds, which deviates somewhat from the values for NaCrO $_2$ and NiGa $_2$ S $_4$ discussed above.

Of course, for the vortex to be a meaningful excitation, minimal amount of noncollinear spin SRO, at least of a few lattice spacings, is required, which, however, is a plausible situation in these compounds in view of the observed spectral broadening of NMR signal and the gapless behavior of the susceptibility.

κ -(BEDT-TTF) $_2$ Cu $_2$ (CN) $_3$ exhibits a dielectric anomaly in the temperature range 20K \sim 40K [35], somewhat above the temperature where the transition-like anomaly is observed in the specific heat or in NMR T_1^{-1} . The AC dielectric constant exhibits a strongly frequency-dependent peak at the time scale of kHz in this temperature range. The observed eminent frequency dependence then suggests that the polarization degree of freedom becomes slowed down significantly in this temperature regime and tends to be frozen. It remains to be seen how this slowing down or freezing of the polarization degree of freedom is related or unrelated to the

spin-liquid and the transition-like behaviors observed at lower temperatures. An extrapolation of the ω -dependent peak temperature to $\omega \rightarrow 0$ yields an estimate of the static dielectric transition temperature $T_c \sim 6\text{K}$, which happens to come close to the specific-heat hump temperature. On the basis of such an observation, Ref.[35] argued that the anomaly of the dielectric constant might be linked to the anomaly of the specific heat [35]. However, such coincidence should be taken with care, since the dielectric measurements are performed already at the macroscopic time scale ($\sim\text{kHz}$ order) not much different from the time scale of thermal measurements and the observed peak temperature of the AC dielectric constant ($20\text{K}\sim 40\text{K}$) is still considerably higher than the specific-heat hump temperature (6K). Thus, at the time scale of real measurements, the specific-heat hump is likely to be a phenomenon not directly related to the dielectric anomaly.

The T -linear specific heat and of the thermal conductivity observed at low temperatures cannot simply be explained by spinwaves. Some other excitations different from spinwaves or any type of Goldstone modes are definitely required here. Two possibilities may be given here: One is a fermionic ‘spinon’ excitation possibly realized in the RVB-like quantum spin-liquid state, as extensively discussed in the literature [60, 61]. The other possibility I wish to propose here is a more conservative one. As discussed above, since the polarization within a molecule has been dynamically frozen at temperatures higher than the transition temperature, and since the spin is likely to be coupled to the polarization in some way or other, randomly frozen polarization might act as a quenched randomness to the spin dynamics, which might give rise to low-energy spin excitations with a constant density of states at low energies, as often encountered in random spin systems. In this context, it might be interesting to point out that Ref.[30] observed that applied magnetic fields induced inhomogeneous magnetization in $\kappa\text{-(BEDT-TTF)}_2\text{Cu}_2(\text{CN})_3$, which was ascribed to impurities or defects. Of course, one needs to explain the reason why the spin itself escapes the freezing in such a situation, in contrast to the standard spin-glass case. Further study is required to clarify the nature of low-energy excitations relevant to the low-temperature properties of these compounds.

Anyway, one should recall that the issue of the low-energy excitations responsible for the low-temperature behavior could more or less be independent of the issue of a finite-temperature transition. Within the Z_2 -vortex ordering picture, in particular, any type of excitation possibly realized in the vortex-free sector in the phase space, can be a candidate of relevant low-energy excitation.

(d) $\text{Cu}_3\text{V}_2\text{O}_7(\text{OH})_2 \cdot 2\text{H}_2\text{O}$

Volborthite $\text{Cu}_3\text{V}_2\text{O}_7(\text{OH})_2 \cdot 2\text{H}_2\text{O}$ is a $S=1/2$ Heisenberg AF on the kagome lattice [62, 63, 64, 65, 66, 67, 68, 69]. In this compound, the lattice is slightly distorted in a uniaxial manner, *i.e.*, a regular triangle being distorted to an isosceles triangle. The extent of the distortion, however, seems not so large. Vesignieite $\text{BaCu}_3\text{V}_2\text{O}_8(\text{OH})_2$ is also a $S=1/2$ kagome-lattice Heisenberg AF with a small lattice distortion [70, 71]. In vesignieite, the lattice distortion is even less than in volborthite.

Volborthite also exhibits a spin-liquid-like behavior in the sense that no AF LRO is observed down to very low temperature [62, 63, 64]. Recent neutron scattering experiment has revealed the onset of the magnetic short-range order at low temperatures [65]. Interestingly, NMR and specific heat measurements have revealed that volborthite exhibits a transition-like anomaly at a finite temperature [68, 62, 63, 64, 72]. Indeed, the NMR relaxation rate T_1^{-1} exhibits a sharp peak suggestive of a thermodynamic phase transition at around $T = T^* = 0.9\text{K}$ [62, 63], while the specific heat shows a hump or a kink at $T_K = 1.05\text{K}$ [64, 72]. Interestingly, a T -linear behavior of the specific heat is observed below T^* together with the T^2 -term, in spite of the fact that volborthite is a very good insulator. Similar behavior, *i.e.*, a combination of the T -linear term and the T^2 term, is observed also above T^* , but with different coefficients from those at $T < T^*$. A closer look at the data reveals that the peak temperature of NMR T_1^{-1} and the kink (hump) temperature of the specific heat are not the same, the former $T = 0.90\text{K}$ being about

15% lower than the latter $T = 1.05\text{K}$. Such $\sim 15\%$ difference reminds us of the T_v vs. T_{peak} relation associated with the Z_2 -vortex ordering.

Below T^* , spins remain fluctuating but the dynamics becomes very slow, as evidenced by NMR T_2^{-1} measurements [63]. (Volborthite also exhibits a weak spin-glass-like freezing phenomenon at a temperature different from T^* , which, however, is of extrinsic origin [62, 63].) This slowly fluctuating low-temperature phase remains stable against weak magnetic fields. When an applied field intensity exceeds a critical value of $\sim 4.3\text{T}$, the system exhibits a phase transition into the more conventional AF long-range ordered state [62, 63].

Vesignieite also exhibits a spin-liquid-like behavior at low temperatures [70, 71]. Although fuller experimental analysis is yet to be done, smaller lattice distortion of vesignieite makes this material a quite attractive model compound.

Many of the above features of volborthite, *i.e.*, [A] the onset of a sharp dynamical anomaly at a finite temperature T^* , [B] the specific-heat hump (kink) temperature located slightly above ($\sim 15\%$) the dynamical transition temperature T^* , [C] appearance of a slowly fluctuating low-temperature phase without the conventional AF nor spin-glass LRO at $T < T^*$, and [D] the fluctuating low-temperature state stabilized against weak applied fields, which is transformed into the conventional LR ordered state upon application of stronger fields, are all quite similar to the features observed in triangular-lattice Heisenberg AFs NaCrO_2 and NiGa_2S_4 discussed above, and are also fully consistent with the Z_2 -vortex ordering picture.

The existence of the T -linear term in the low-temperature specific heat is a feature absent in NaCrO_2 and NiGa_2S_4 (though present in organic triangular AFs). The behavior of the low-temperature specific heat reflects the properties of the low-energy excitation of the system. If the relevant low-energy excitations are Goldstone modes like spinwaves, a T^2 term should arise as is observed in NaCrO_2 and NiGa_2S_4 . Hence, the existence of the T -linear term indicates that the relevant low-energy excitation in volborthite is exotic. Indeed, Kagome AF has been known to sustain exotic localized low-energy excitations like the weathervane excitation, which might lead to the observed T -linear specific heat.

It should be noticed again, however, that the properties of the low-temperature phase, *e.g.*, the existence or nonexistence of the T -linear term in the low-temperature specific heat, is a property governed by the non-vortex physics, an issue independent of the Z_2 -vortex transition itself. In other words, the Z_2 -vortex ordering scenario is well compatible with various different scenarios of the low-temperature phase, as long as the relevant low-energy excitations are realizable in a restricted vortex-free (topologically simply-connected) phase space.

The other aspect to be considered is the role played by the Dzaloshinskii-Moriya (DM) interaction, which inevitably exists to certain extent in any kagome-lattice magnet. The DM interaction tends to lower the symmetry, lift the degeneracy of states, and thereby favor the ordering (usually favors the $q = 0$ state). Thus, its effect on the nature of low-energy excitations needs to be further examined.

(e) $\text{SrCr}_{8-x}\text{Ga}_{4+x}\text{O}_{19}$

$\text{SrCr}_{8-x}\text{Ga}_{4+x}\text{O}_{19}$ (SCGO) is a $S=3/2$ kagome-bilayer (or pyrochlore slab) system, extensively studied in 1990's as a typical model system of kagome-lattice Heisenberg AF [73, 74, 75, 76, 77, 78, 79, 80, 81, 82, 83]. Crystal structure of magnetic ions (Cr^{3+}) in this material, however, is not really a planar kagome, but rather a kagome-bilayer where two kagome layers are connected via a sparse triangular layer in between, *i.e.*, a pyrochlore slab along (111) [84, 85]. Thus, a constituent unit of the lattice is a tetrahedron rather than a triangle. Another point to be noticed is that a certain fraction of Cr ions are randomly diluted by nonmagnetic Ga so that the lattice contains sizable amount of disorder, which might affect the magnetic ordering of this material to some extent.

SCGO exhibits a rather sharp spin-glass-like transition at a finite temperature T_f characterized by a cusp in the susceptibility accompanied by the deviation between the FC and

ZFC data. Even the negative divergence of the nonlinear susceptibility, which is a characteristic of the standard spin-glass order, was observed [73]. This spin-glass-like transition, however, is unusual in the sense that (i) only a fraction of spin is frozen in the low-temperature phase and large spin fluctuations remain even below T_f , (ii) the low-temperature specific heat is proportional to T^2 suggestive of 2D spinwaves (or certain Goldstone modes) in sharp contrast to the standard spin-glass case where the low-temperature specific heat exhibits a T -linear behavior, and (iii) the T_f -value of SCGO tends to increase with decreasing the defect concentration, apparently suggesting that the defect might not be essential in the spin-glass-like transition of SCGO [82]. The specific heat exhibits a rounded peak slightly above T_f without an appreciable anomaly at $T = T_f$, as commonly observed in standard spin glasses. Very much similar behavior was also reported for other $S=3/2$ kagome-bilayer compound $\text{Ba}_2\text{Sn}_2\text{ZnCr}_{7p}\text{Ga}_{10-7p}\text{O}_{22}$ (BSZCGO) [86, 87, 88, 89].

Some of the above features of SCGO, *e.g.*, considerable fluctuations remaining in the ordered phase and the specific-heat peak slightly above T_f , seem common with the features observed in other frustrated magnets discussed above, NaCrO_2 , NiGa_2S_4 and volborthite. The occurrence of a clear spin-glass transition in SCGO, accompanied by the divergence of the nonlinear susceptibility, is at odds with other magnets, however. The implication from the observation (iii) above might be the spin-glass transition of SCGO is not intrinsically defect-mediated.

One possibility in the context of the Z_2 -vortex ordering scenario might be that the spin-glass transition of SCGO is essentially a Z_2 -vortex transition, which takes a character of the spin-glass transition due to the presence of significant amount of defects (randomness). In other words, the Z_2 -vortex transition serves as a “seed” of the experimentally observed spin-glass transition. In view of the fact that the observed spin-glass transition is quite pronounced accompanying the associated critical phenomena, it remains to be seen what role the Z_2 -vortex is playing here. The true nature of the spin-glass transition of SCGO has remained elusive yet.

4. Summary and discussion

We discussed the recent experimental data on various frustrated quasi-2D Heisenberg AFs from the viewpoint of the Z_2 -vortex order, which include (a) $S=3/2$ triangular-lattice AF NaCrO_2 , (b) $S=1$ triangular-lattice AF NiGa_2S_4 , (c) $S=1/2$ organic triangular-lattice AFs κ -(BEDT-TTF) $_2\text{Cu}_2(\text{CN})_3$ and $\text{EtMe}_3\text{Sb}[\text{Pd}(\text{dmit})_2]_2$, (d) $S=1/2$ kagome-lattice AF volborthite $\text{Cu}_3\text{V}_2\text{O}_7(\text{OH})_2 \cdot 2\text{H}_2\text{O}$, and (e) $S=3/2$ kagome-bilayer AF $\text{SrCr}_{8-x}\text{Ga}_{4+x}\text{O}_{19}$.

These magnets exhibit the spin-liquid-like behavior without the conventional AF LRO at low temperatures. Furthermore, most of them exhibit the following features in common: [A] onset of a sharp dynamical anomaly at a finite temperature T^* without a strong anomaly in the specific heat, [B] the specific-heat exhibiting a hump (or a kink) slightly above the dynamical transition temperature T^* , [C] appearance of a slowly fluctuating low-temperature state without the conventional AF nor spin-glass order at $T < T^*$, and [D] the fluctuating low-temperature state stabilized against weak applied fields, which is transformed into the conventional LR ordered state upon application of stronger fields. These features are observed in common at least for [a] NaCrO_2 , [b] NiGa_2S_4 and [d] volborthite. Note that these materials are already of quite a variety, either $S=3/2$, 1 or $1/2$, and either triangular or (distorted) kagome. Hence, the above features [A]-[D] should possess universal origin independent of the spin quantum number and the detailed lattice structure. The Z_2 -vortex ordering scenario meets such a universal criterion since the conditions required there are, (i) the system should be 2D, *i.e.*, weak enough interplane coupling, (ii) the system should be Heisenberg-like, *i.e.*, weak enough magnetic anisotropy, (iii) the existence of the noncollinear spin SRO arising from frustration. All compounds discussed above satisfy these criteria to certain extent, and could be candidates of the Z_2 -vortex bearing system. Such universal features of the Z_2 -vortex ordering scenario is a favorable factor in explaining why the exotic properties [A]-[D] are observed in common in a variety of 2D frustrated

Heisenberg AFs with different lattice geometries and spin quantum numbers.

$S=1/2$ organic compounds κ -(BEDT-TTF) $_2$ Cu $_2$ (CN) $_3$ and EtMe $_3$ Sb[Pd(dmit) $_2$] $_2$ exhibit somewhat different properties from NaCrO $_2$, NiGa $_2$ S $_4$ and volborthite discussed above. For example, T_{peak}/T^* -value takes a larger value of ~ 2 . Dynamical slowing down in the low-temperature phase below T^* seems less pronounced as compared with the one in the other compounds. As discussed in §3(c), the latter property as well as the T -linear low-temperature specific heat might reflect a novel type of low-energy excitations inherent to these compounds, but may well be compatible with the Z_2 -vortex order. Even for $S=3/2$ bilayer-kagome magnet SCGO, Z_2 -vortex might be playing some role. The open issue here is to clarify the nature of the observed spin-glass-like transition.

Overall, the Z_2 -vortex ordering scenario seems to give consistent account of a number of exotic ordering properties observed in a variety of frustrated quasi-2D Heisenberg magnets in common. Hopefully, further experimental and theoretical studies such as inelastic neutron-scattering will reveal further novel features of the Z_2 -vortex and other exotic excitations inherent to frustrated magnets.

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